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Exciton-hole collision in staggered type-II $Al_{0.34}Ga_{0.66}As/AlAs$ multiple quantum wells

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We report the phase relaxation of Γ-Γ direct excitons in staggered type-II $Al_{0.34}Ga_{0.66}As/AlAs$ multiple quantum wells by means of the time-resolved degenerate four-wave mixing. The dephasing rate increases with the increase in the excitation density. Its excitation density dependence sharply changes at a critical density of 0.2 $\mu J/cm^2$. The dependence agrees with the excitation density dependence of the long-lived Γ-hole density studied by means of the pump-and-probe experiment. The long-lived hole density saturates at the same excitation density of 0.2 $\mu J/cm^2$. These facts definitely indicate that the phase relaxation rate of Γ-Γ excitons is dominated by the collision between the excitons and long-lived holes.

Recently, active studies have been carried out on carrier scattering processes in semiconductors by means of ultrafast spectroscopy.[1-3] Above all, the carrier-carrier scattering process in the semiconductor confined system is particularly interesting, because new kinds of boundary conditions are given to the carrier scattering.[2-6] In type-I quantum wells where electrons, holes, and excitons are confined in the same well layer, the collision rates of an exciton with an incoherent exciton and a free carrier (an electron and a hole) were extensively studied. The exciton–free-carrier collision rate was found to be about 8 times larger than the exciton-exciton collision rate.[3,6]

On the other hand, the exciton collision mechanism in the staggered type-II system has not been well studied yet.[7] In staggered type-II $Al_xGa_{1-x}As/AlAs$ multiple quantum wells, the conditions are very unique as a result of the interlayer Γ-X scattering process.[8-10] The main feature of the carrier dynamics in the staggered type-II multiple quantum wells is characterized by the rapid Γ-X interlayer scattering and the slow indirect Γ-X recombination.[9-13] The Γ-electron state in the $Al_xGa_{1-x}As$ well layer is located at higher energy than the X-electron state in the AlAs barrier layer. Thus, photoexcited Γ electrons in the well layer are quickly scattered into the X state in the barrier layer within a few picoseconds.[7,9,10-13] The recombination process of Γ holes in the well layer and X electrons in the barrier layer takes place in a microsecond time scale, because of the indirect nature in both the real and momentum space.[10,14-16] The long-lived Γ holes may contribute to the phase relaxation of the Γ-Γ direct excitons in the well.

In this work, we observed the exciton collision process in staggered type-II multiple quantum wells by means of the time-resolved degenerate four-wave mixing. The phase relaxation rate of the Γ-Γ direct excitons increases with the increase in the excitation density and the repetition rates of laser pulses. The excitation density dependence of the phase relaxation rate is represented by a couple of straight lines with a bend at a certain excitation density. The dependence is the same as the excitation density dependence of the long-lived hole density measured by the pump-and-probe transient absorption experiment. The bends are attributable to the saturation of the long-lived hole density. These facts definitely indicate that the collision of the excitons with the long-lived holes generated by the preceding laser pulses is the major collision mechanism.

The samples studied in this experiment were 100 alternate layers of 9.2-nm $Al_{0.34}Ga_{0.66}As$ and 2.7-nm AlAs. The sample was directly immersed in superfluid liquid helium at 2 K. Absorption and luminescence spectra of the sample are shown in our previous paper.[8,10] The lowest-energy heavy-hole exciton peaks at 2.034 eV and is inhomogeneously broadened. The time-resolved degenerate-four-wave-mixing experiment was carried out by using a subpicosecond cavity-dumped dye laser. The photon energy of the laser pulses was resonantly tuned to the absorption peak position of the lowest exciton, 2.034 eV. The spectral width and the temporal width of the laser pulses were about 6 meV and 500 fs, respectively. It is known that the time resolution of the time-resolved four-wave mixing is determined by the inverse of the spectral width.[17] Therefore, our time resolution is about 300 fs corresponding to the laser spectral width of 6 meV. To avoid experimental difficulties keeping the good signal-to-noise ratio, we used the two-pulse configuration in the standard forward-scattering geometry. The arrangement of the experiment is displayed in the inset of Fig. 1. Two laser pulses propagating in the direction of $\mathbf{k}_1$ and $\mathbf{k}_2$, which are parallel polarized to each other, interfere in the sample and generate a grating as long as their time delay $\tau=t_2-t_1$ is of the order of the exciton dephasing time T_2. The second pulse was diffracted by the grating and the signal was generated in the direction $\mathbf{k}_3=2\mathbf{k}_2-\mathbf{k}_1$. In the inhomogeneously broadened two-level system, the optical coherence time T_2 of the system is given by 4τ, where τ is the decay time constant of the time-resolved degenerate-four-wave-mixing signal.[18] In this way, we obtained T_2 of the excitons by measuring the decay time constant τ. The coherence relaxation rate of excitons is an inverse of T_2. When the T_2 is dominated by the lifetime T_1, T_2 is given by $2T_1$. The intensity

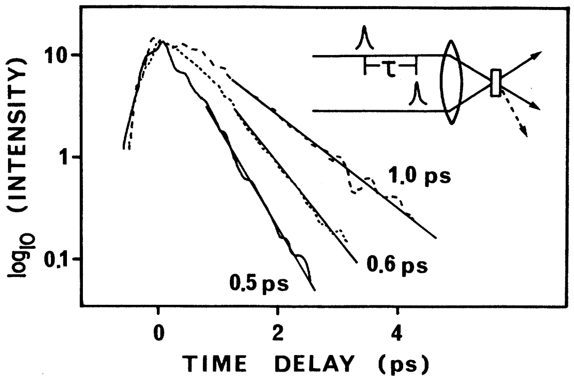

FIG. 1. Typical time traces of the degenerate four-wave mixing at the Γ-Γ exciton transition in $Al_{0.34}Ga_{0.66}As/AlAs$ multiple quantum wells. The repetition rate of the laser pulses is 4.1 MHz. Traces shown by a dashed line, a dotted line, and a solid line correspond to three excitation densities 0.07, 0.22, and 0.63 $\mu J/cm^2$, respectively. Note that the decay time constant becomes short with the increase in the excitation density.

of the second refocusing pulse was always kept at the quarter of the first pulse intensity. The intensities of the two pulses were changed simultaneously. Hereafter, we mean that the excitation density is that of the first pulse.

The typical time traces of the degenerate four-wave mixing for three excitation densities are shown in Fig. 1. The repetition rate of the laser pulses is 4.1 MHz. The excitation densities shown in the figure caption are those of the first pulse. The decay of the signal is single exponential decay. The best fits of the decay curves give the decay time constant of 1.0, 0.6, and 0.5 ps for the traces corresponding to the densities of 0.07, 0.22, and 0.63 $\mu J/cm^2$, respectively. The decay time becomes shorter as the excitation density increases.

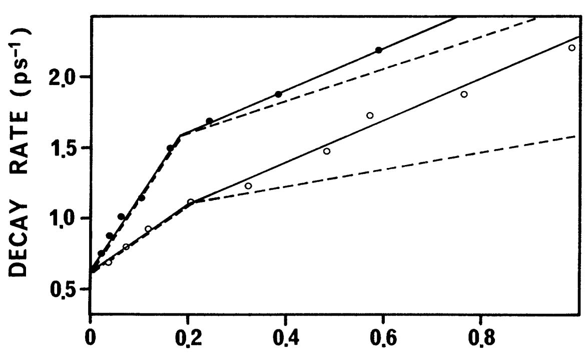

FIG. 2. The plot of the decay rate $1/\tau$ of the degenerate-four-wave-mixing signal vs the excitation density. The vertical scale is equal to $4/T_2$. Closed and open circles correspond to the pulse repetition rates of 4.1 and 0.82 MHz, respectively. There are bends around the excitation density of 0.2 $\mu J/cm^2$. Note that the slopes of the two solid lines are different below the bends and become equal to each other above the bends. Dashed lines are the contribution of the exciton–"old"-hole collision described in the text.

The plots of the decay rate of the signal $1/\tau$ versus the excitation density are displayed in Fig. 2. Two kinds of marks show the data taken at two kinds of repetition rates, 4.1 and 0.82 MHz. They agree with each other at the low-density limit. The low-density limit of the relaxation time is 1.6 ps, which is close to twice that of the Γ-X interlayer scattering time of 1.2 ps.[9] The excitation density dependence of the relaxation rate is represented by a couple of straight solid lines. There are bends around the excitation density of 0.2 $\mu J/cm^2$. Below the bends, the slopes for the two repetition rates are different from each other, while they are equal to each other above the bends. At larger excitation intensity than 0.6 $\mu J/cm^2$, the decay curves of the signal seem to show double exponential decays. In this region, we adopt tentatively the first rapid component as the decay constant. The slow component grows as the excitation density is raised.

To inspect the temporal change of carrier density and its kind, we performed the pump-and-probe transient absorption experiment using the same experimental setup that is used for the time-resolved four-wave mixing. The details of the transient absorption experiment are given in our previous paper.[10] The typical time trace of the transient absorption is shown in Fig. 3. The observed bleaching is attributable to the hole state saturation except at the time origin.[10] The base appearing at the negative time delay is due to the long recombination lifetime of

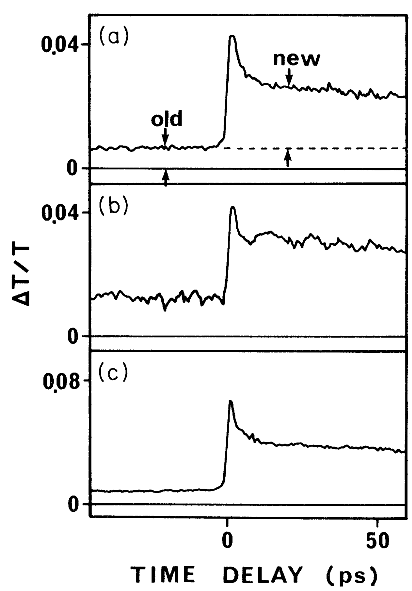

FIG. 3. Typical time traces of the absorption bleaching of the Γ-Γ direct exciton. The energy densities corresponding to (a), (b), and (c) are 0.33, 0.31, and 0.57 $\mu J/cm^2$, respectively. The repetition rates of laser pulses used for (a), (b), and (c) are 0.82, 4.1, and 0.82 MHz, respectively. These bleachings are due to the hole-state saturation and are proportional to the hole density in the well layer as well as the X-electron density in the barrier layer. In (a), "old" and "new" are used to identify the saturation due to the long-lived holes and the saturation due to the holes generated by a single excitation pulse. The rapid decay around the time origin is caused by the fast Γ-X interlayer scattering of electrons.

the X-Γ electron-hole pairs. The lifetime is longer than the present pulse-repetition intervals, 240 ns and 1.2 μs. It is not ascribed to the Γ-Γ exciton population, because the lifetime of the Γ-Γ exciton is less than 10 ps. Therefore, the base is proportional to the unrecombined long-lived hole density created by the preceding pulses.[10] We call the long-lived hole the "old" hole. The transient absorption shows a peak and a fast decay around the zero time delay. The peak is simply ascribed to the Γ-Γ exciton bleaching due to the creation of the Γ-Γ excitons. The fast decay corresponds to the interlayer Γ-X scattering of electrons.[9,10] The transient absorption shows a plateau after the fast decay finishes. It is ascribed to the newly created hole density generated as a result of the dissociation of excitons and the interlayer Γ-X scattering of electrons. We call these holes "new" holes.

Figure 3 shows that the "old" hole density increases with the increase in the excitation density and the repetition rate of laser pulses. The plot of the "old" hole density and the "new" hole density versus the excitation density is depicted in Fig. 4. Again, we plotted the data taken at two kinds of laser repetition rates, 4.1 and 0.82 MHz. With the increase in the excitation density, the "old" hole density increases with sharp bends around the critical excitation density of 0.2 $\mu J/cm^2$. The "old" hole density almost saturates under the excitation density of 0.2 $\mu J/cm^2$. The saturated value for the 4.1-MHz repetition rate is larger than that for the 0.82-MHz repetition rate. The difference is ascribed to the slow recombination of Γ holes and X electrons. The slopes are different below the critical density. The difference means that the "old" hole density decreases during 960 ns, which is equal to 1.2 μs$-$240 ns. On the other hand, the "new" hole density increases nearly in proportion to the excitation density regardless of the laser repetition rate.

Excitation intensity dependence of the dephasing rate of Γ-Γ excitons and that of the "old" hole density are well related to each other. The change appears at the same excitation density of 0.2 $\mu J/cm^2$, as shown in Figs. 2 and 4. The similar excitation density dependence definitely indicates that the excitons dephase mainly through the collision between excitons and "old" holes. As a result of the interlayer Γ-X scattering, Γ-Γ excitons are dissociated into the X electrons in the barrier layer and the Γ holes in the well layer. The Γ holes are alive for a long time. They collide strongly with newly generated excitons. This is the main dephasing mechanism of the Γ-Γ excitons. One may consider that "new" holes should contribute to the exciton-hole collision process. However, "new" holes are created as a result of the dissociation of excitons. Therefore "new" holes have little chance to collide with excitons.

The bends that appeared around the excitation density of 0.2 $\mu J/cm^2$ are explained by the saturation of the density of long-lived holes. The saturation of the long-lived "old" holes does not mean the saturation of the X-state or the inhibition of the Γ-X scattering, because "new" holes do not saturate. The recombination of Γ holes and X electrons is nonexponential, and some electron-hole pairs can recombine rapidly depending on the pair density. Therefore, the long-lived "old" hole density can be saturated without the saturation of "new" hole density. The density-dependent carrier diffusion may also contribute to the saturation of the long-lived "old" hole density.

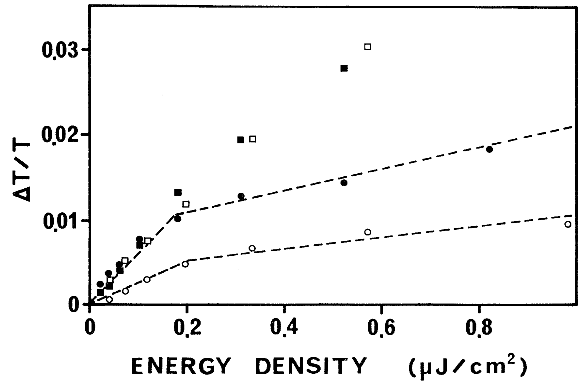

FIG. 4. The plot of the bleaching ascribed to the hole density vs the excitation density. Closed and open circles show the bleaching due to the "old" holes and correspond to the pulse repetition rates of 4.1 and 0.82 MHz, respectively. Closed and open squares are the saturations due to the "new" holes and correspond to the pulse repetition rates of 4.1 and 0.82 MHz, respectively. There is a saturation point for the "old" hole density around the excitation density of 0.2 $\mu J/cm^2$.

It is useful to compare our result for the type-II system with the result for the type-I system. The experimental result reported by Honold *et al.* for a type-I GaAs single quantum well is as follows.[6] The excitation density dependence of the dephasing rate of the lowest-energy exciton is determined by the exciton–free-carrier and exciton-exciton collisions. The dephasing rate is written by $1/T_2 = A + B\ N$, where A is a constant independent of the carrier density, N is a density of excitons or free carriers in units of $10^{10}\ cm^{-2}$, and B is 0.087 ps^{-1} for the exciton-exciton collision or 0.67 ps^{-1} for the exciton–free-carrier collision. To compare our result with the result mentioned above, we estimated the created exciton density. The excitation density of 1 $\mu J/cm^2$ corresponds to the sheet exciton density of 2.6×10^{10} cm^{-2} by using the optical density of 0.82 at the exciton absorption peak. Figure 4 informs us of the excitation density dependence of "old" and "new" hole densities, because an exciton is converted to a "new" hole and because a "new" hole and an "old" hole contribute to the same amount of absorption bleaching. Using the value of $B = 0.51\ ps^{-1}$ for the exciton–"old"-hole collision, we estimated the contribution of the exciton–"old"-hole collision to the observed decay rate, shown by dashed lines in Fig. 2. We can explain the data of Fig. 2. The value of B for the exciton–"old"-hole collision almost agrees with that for the exciton–free-carrier collision in the type-I system. However, the additional collision mechanism must be taken into account above the bends. The value of B for the additional collision is 0.033–0.084 ps^{-1}. It is probably the exciton-exciton collision or the exciton–"new"-hole collision. If the exciton-exciton collision works, the value of B almost agrees with that for the exciton-exciton collision in the type-I system. The growth of the slow component in the degenerate-four-wave-mixing signal under the higher excitation density may be explained in this picture.

In conclusion, we studied the exciton collision processes in type-II multiple quantum wells by means of the time-resolved four-wave mixing and the pump-and-probe transient absorption. The exciton dephasing rate increases with the increase in the excitation density and the repetition rate of laser pulses. We found that the collision rate of the Γ-Γ exciton strongly depends on the dissociated long-lived hole density created by the preceding laser pulses. This result indicates that the dephasing of the Γ-Γ exciton is dominated by the collision between excitons and long-lived holes.

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